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HIGH RESOLUTION LASER SPECTROSCOPY OF COLOUR CENTRES
(U) IBM RESEARCH LAB SAN JOSE CA R M MACFARLANE ET AL.
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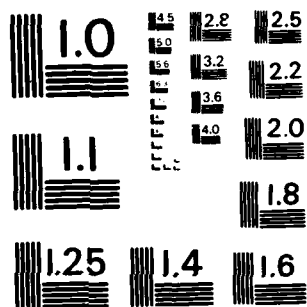
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TECHNICAL REPORT NO. 4

High Resolution Laser Spectroscopy

of Colour Centres

by

R. M. MacFarlane, R. T. Harley, and R. M. Shelby

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INTRODUCTION

The spectroscopy of zero-phonon lines has probably been the most important source of information about the symmetry and nature of aggregate colour centres in solids.¹ The usefulness of these lines as probes of defect centres depends on their having a linewidth narrow enough to resolve shifts and splittings due to external perturbations such as magnetic and electric fields and uniaxial stress. At liquid helium temperatures, the widths of zero phonon lines are substantially less than at higher temperatures, but they are still inhomogeneously broadened by lattice strains, and a residual width (Γ_{inh}) of 1-50 cm^{-1} remains. This can prevent the observation of resolved splittings and necessitate the use of modulation techniques² and moment analysis³ to extract any useful information.

As will be discussed here, the use of narrow band laser excitation makes it possible to eliminate the effects of this inhomogeneous broadening and realize a resolution limited only by the homogeneous linewidth (Γ_h) which is typically 10-100 MHz. This increase in resolution by a factor of 10^3 - 10^4 over conventional spectroscopy impacts at least two areas. The first is high resolution studies of the effect of external perturbations and hence the physical and electronic structure of the centres; and the second is a determination of the mechanisms responsible for homogeneous broadening or equivalently the optical dephasing time $T_2 = (\pi\Gamma_h)^{-1}$. Contributions to T_2 come from population decay (T_1) as well as from pure dephasing (T_2^*) due to random modulation of the optical transition frequency by phonons, nuclear spins, etc., i.e., $(T_2)^{-1} = (2T_1)^{-1} + (T_2^*)^{-1}$. Considerable work is being done in this field to study rare earth impurity systems in crystals⁴ and glasses.⁵ Here we concentrate on the results that have been obtained from the application of these techniques to colour centre systems which exhibit zero phonon lines. The techniques are basically of

two kinds—frequency domain methods such as holeburning and time domain observations of optical coherent transients.

HOLEBURNING

Holeburning is the reduction of absorption following selective narrow band laser excitation within the inhomogeneous line, (see Figure 1). Holeburning due to two-level saturation was observed for the R lines in ruby.⁶ Such holes recover in the absence of exciting light with the excited state lifetime. Longer lived holes were observed for organic molecules in crystals⁷ due to selective photochemistry and in glasses⁸ due to rearrangement of molecular environments. Soon after this, optical pumping of the nuclear hyperfine levels of rare earth ions in crystals was shown to lead to holeburning⁹ with the hole lifetime being determined by nuclear spin-lattice relaxation times, which varied from ~secs¹⁰ to ~1 hr.¹¹ The first observation of holeburning in a colour centre material was reported by Macfarlane and Shelby¹² for the F_3^+ centre in NaF, and since then experiments have been carried out in a number of other colour centre systems (see Table I), suggesting that the phenomenon of holeburning in colour centres may be quite general. In addition, there are possible applications to information storage.¹³

Two cases have been investigated in some detail, i.e., F_3^+ in NaF¹² and the 6070A zero phonon line in NaF.^{14,15}

a) F_3^+ in NaF

This centre is a singly ionized aggregate of three F centres in a (111) plane having C_{3v} symmetry. The emission spectrum ($^1E \rightarrow ^1A_1$) exhibits a zero phonon line at 5456A¹⁶ and approximately mirrors the absorption (see Figure 2). The zero phonon line has $\sim 10^{-3}$ of the intensity of the total transition and an inhomogeneous width at 2K of 1.1 cm^{-1} (32 GHz).

Holeburning was observed by irradiating the zero phonon line for ~ 2 sec with 200 mW/cm^2 of single frequency dye laser light with a jitter width (Γ_j) of ~ 2 MHz. The resulting holes were detected by scanning the laser at reduced intensity and measuring the excitation spectrum of ${}^1E \rightarrow {}^1A_1$ emission in the peak of the phonon side band at 5800 Å (see Figure 2). The hole width, H , was 38 MHz and since the hole depth was $< 10\%$ the homogeneous width was obtained from $H = 2(\Gamma_h + \Gamma_j)$ i.e., $\Gamma_h = 17$ MHz or $T_2 = 20$ nsec. The fluorescence decay time, T_1 , of this centre is 10 nsec. The homogeneous linewidth is thus T_1 -limited, a situation that is expected to be common for strongly allowed transitions.

The hole recovery showed two components, one of several seconds due to a triplet state bottleneck, and a much longer component of 70 minutes (see Figure 3). The long-lived holeburning could be reversed by irradiation with near UV light, with an action spectrum which followed approximately the F band absorption which peaks at 335 nm. This suggests that an electron is lost from the centre to a trap during holeburning and can be replaced by electrons liberated by the UV light. The maximum hole depth which could be obtained was $\sim 30\%$, indicating that not all F_3^+ centres were sufficiently near appropriate electron traps.

The observation of long-lived narrow holes suggested that an enormous increase in resolution could be achieved in Stark spectroscopy since a d.c. electric field could be applied and the holes scanned during their long lifetime. The electric field removes the electronic degeneracy of the 1E state with a Stark splitting coefficient A , as well as the orientational degeneracy (due to the cubic crystal symmetry) with a pseudo-Stark coefficient B . For electric fields (E_s) parallel to (100) a distribution of splittings was found peaking at $|A - B| = 0.19 \text{ MHz/Vcm}^{-1}$ for laser polarization E_L parallel to E_s and $|A + B| = 0.43 \text{ MHz/Vcm}^{-1}$ for $E_L \perp E_s$ (see Figure 4). The distribution of intensities results from the distribution of magnitudes and orientations of local strain fields with respect

to the applied electric field. This provides clear evidence that there exists a distribution of zero-field splittings of the E level inside the inhomogeneous line. The splittings will be on the order of the inhomogeneous line-width, since the coefficients for stress-induced removal of the electronic degeneracy are comparable to those for removal of orientational degeneracy. Since the strain fields strongly influence only the Stark splitting, the latter can be distinguished from the pseudo-Stark term by an analysis of the intensity distribution of Figures 4(b) and 4(c). Such an analysis shows that for (100) fields, the Stark-splitting term ($A=0.31 \text{ MHz/Vcm}^{-1}$) is larger than the pseudo-Stark term ($B=0.12 \text{ MHz/Vcm}^{-1}$). The latter represents twice the difference between ground and excited state shifts which is the difference of the projections along (100) of the ground and excited state (111) expectation dipole moments. The magnitude of this difference, $|\mathbf{d}_1^e - \mathbf{d}_1^g|$, is $(\sqrt{3}/2) B = 0.10 \text{ MHz/Vcm}^{-1}$, expressed in energy splitting units. The true Stark effect measured the dipole moment perpendicular to the 3-fold axis i.e., $\mathbf{d}_1^e = (\sqrt{3}/2\sqrt{2}) A = 0.19 \text{ MHz/Vcm}^{-1}$.

The Zeeman effect was also investigated by burning a hole and then applying magnetic fields up to 50 kG. From magnetic-circular-dichroism measurements, Davis and Fitchen¹⁷ obtained a g-value of 0.04 ± 0.02 for this transition. Thus a linear splitting of up to 2.8 GHz was expected. However, the magnetic field had no observable effect on the hole. This provided further evidence that sizeable zero-field splittings due to random internal strains are present and quench the linear Zeeman effect. An analysis of the Zeeman effect in the presence of a distribution of zero-field splittings shows that the magnetic interaction between the split components of 1E is certainly less than $0.05\mu_B$. This is consistent with the magnetic-circular-dichroism results,¹⁷ which cannot detect the presence of zero-field splittings.

of m and ℓ quoted above. An example of a set of experimental data is shown in Figure 6 for $E_s \parallel (111)$. In this centre, the ground and excited states are nondegenerate ($A' \leftrightarrow A''$) so the electric field splittings are pseudo-Stark splittings i.e., arise from the removal of orientational degeneracy only. The holes, therefore, do not broaden significantly in the applied field. The three sets of splittings in Figure 6 are in the ratio $|2m - \ell| : \ell : (2m + \ell)$ from which $\ell/m = 1.446$ is obtained, with the magnitude of the dipole being $1.57 \text{ MHz/Vcm}^{-1}$.

c) Other Centres

In addition to the two cases discussed above, holeburning has been found in a number of other centres (see Table I). So far, studies have concentrated on spectral regions easily accessible to cw dye lasers and a large fraction of the centres studied exhibit holeburning. In most cases, those which fail to "burn" do not fluoresce i.e., they are quenched by fast nonradiative relaxation and have a broad homogeneous linewidth.

COHERENT TRANSIENTS

Photon echo or optical free decay techniques are ideally suited to measuring homogeneous linewidths because the measurements are made on the time scale of the dephasing itself. They are therefore less sensitive to the effects of slowly changing environments or slow spectral diffusion which can lead to line broadening in frequency domain techniques such as holeburning. Two factors are probably responsible for the lack of work in this area on colour centre materials. One is the expectation that in most cases at the lowest temperatures the dephasing time will be limited by the fluorescence lifetime which can be more easily measured in other ways, and the second is the widespread occurrence of long lived holeburning which leads to substantial bleaching of the zero phonon line at the high intensities used in coherent transient measurements.

A measurement of optical free induction decay in the F_3^+ centre of NaF²⁰ using laser frequency switching by an intracavity phase modulator²¹ gave a dephasing time of $T_2 = 16 \pm 4$ nsecs. This is consistent with the holeburning ($\Gamma_h = 17$ MHz) and fluorescence lifetime measurements ($T_1 = 10 \pm 2$ nsec) and confirms that optical dephasing is due to population decay. For these fast dephasing times the technique of optical phase switching²² is more easily applied. Here a coherent superposition of ground and excited states is produced in steady state and then the phase of the laser is switched by applying a voltage step to a phase modulator outside the laser cavity. The coherent sample polarization radiates in the forward direction and the laser acts as a local oscillator for homodyne detection. Maximum signal strength occurs for a phase switch of π radians. It has been shown²² that the signal decay is simply related to T_1 and T_2 in the low power regime i.e., $\chi^2 T_1 T_2 < 1$ where χ is the optical Rabi frequency. In particular, the phase switch signal decays exponentially with a rate $2/T_2$. Phase switched decays were observed on the 6070A line in NaF giving $T_2 = 10 \pm 2$ nsecs in agreement with the holeburning result (see Figure 7a). Measurements were also made on the 5754A centre and the decays here were too fast to be resolved by our detection system i.e., $T_2 \leq 2$ nsecs (Fig. 7b). In these experiments, the laser was gated on for 200 μ sec during the phase switching and the repetition rate of the experiment was 50 Hz. This reduced the effects of long lived holeburning.

CONCLUSION

The examples discussed in this paper serve to illustrate the usefulness of holeburning for very high resolution spectroscopic studies of colour centres, as well as for obtaining a measure of the homogeneous linewidths i.e., optical dephasing times. It is expected that the technique will become a rather generally useful tool for such studies. Coherent transient techniques have not yet been applied in many cases but examples of optical phase switched

decays and optical free induction decay show promise of future applications. Photon echo measurements using picosecond pulses have not yet been made but should prove particularly useful for the determination of dephasing times.

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Table I. Colour Centres Exhibiting Long Lived Holeburning

Material	$\lambda(\text{\AA})^\dagger$	$\Gamma_{\text{inh}}(\text{GHz})$	Assignment	Ref.
			or Symmetry	
NaF	5455.50	32	F_3^+ , C_{3v}	a,b
	5769.30	40	—	c,d
	5753.65	34	—	c
	6069.65	36	C_S	e,f
LiF	8330	130	R' , C_{3v}	g
CaF_2	6774	130	F_3 , D_{2d}	c
Diamond	6378	900	vac-N pair, C_{3v}	c

† Wavelength (in air) of the zero phonon line.

a. R. M. Macfarlane and R. M. Shelby, Phys. Rev. Lett. 42, 788 (1979).

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FIGURE CAPTIONS

Figure 1. Schematic illustration of an inhomogeneously broadened line with a hole bleached at the frequency of a narrow band laser. For shallow holes, the hole width is twice the homogeneous linewidth.

Figure 2. (a) Schematic energy-level diagram for the ${}^1A_1 \leftrightarrow {}^1E$ transition of F_3^+ in NaF; (b) the emission of the zero-phonon line and the vibronic sideband; (c) the excitation spectrum; (d) a hole burnt in the zero-phonon line.

Figure 3. Hole recovery time for the 5456A line of F_3^+ in NaF. (a) The slow component and (b) the fast triplet population component.

Figure 4. Stark effect in F_3^+ (a) Hole burned in zero field. (b),(c) Applied field $E_s = 2.7 \text{ kV/cm}$. $\parallel (100)$. E_L denotes the direction of the laser polarization. A is the pseudo-Stark coefficient and B the Stark coefficient.

Figure 5. Stark splitting patterns for the C_s centre at 6070A in NaF. E_s denotes the Stark field and E_L the laser polarization used for burning and probing. The direction of the permanent dipole is (m,m,l'). On the left side of the figure theoretical intensities are given, and on the right experimental intensities are shown in parenthesis. Note that the experimental values have been normalized to one theoretical value for each of the 7 experimental geometries.

Figure 6. Stark effect on the 6070A centre in NaF using holeburning spectroscopy. The case $E_s \parallel (111)$ is shown here (a) $E_L \parallel (111)$, (b) $E_L \parallel (110)$, (c) Stark splittings as a function of applied electric field.

Figure 7. Measurement of optical dephasing by phase switching. (a) 6070A centre in NaF, (b) 5754A centre in NaF. In case (b), the decay time is limited by the detector response and is < 2 nsec.

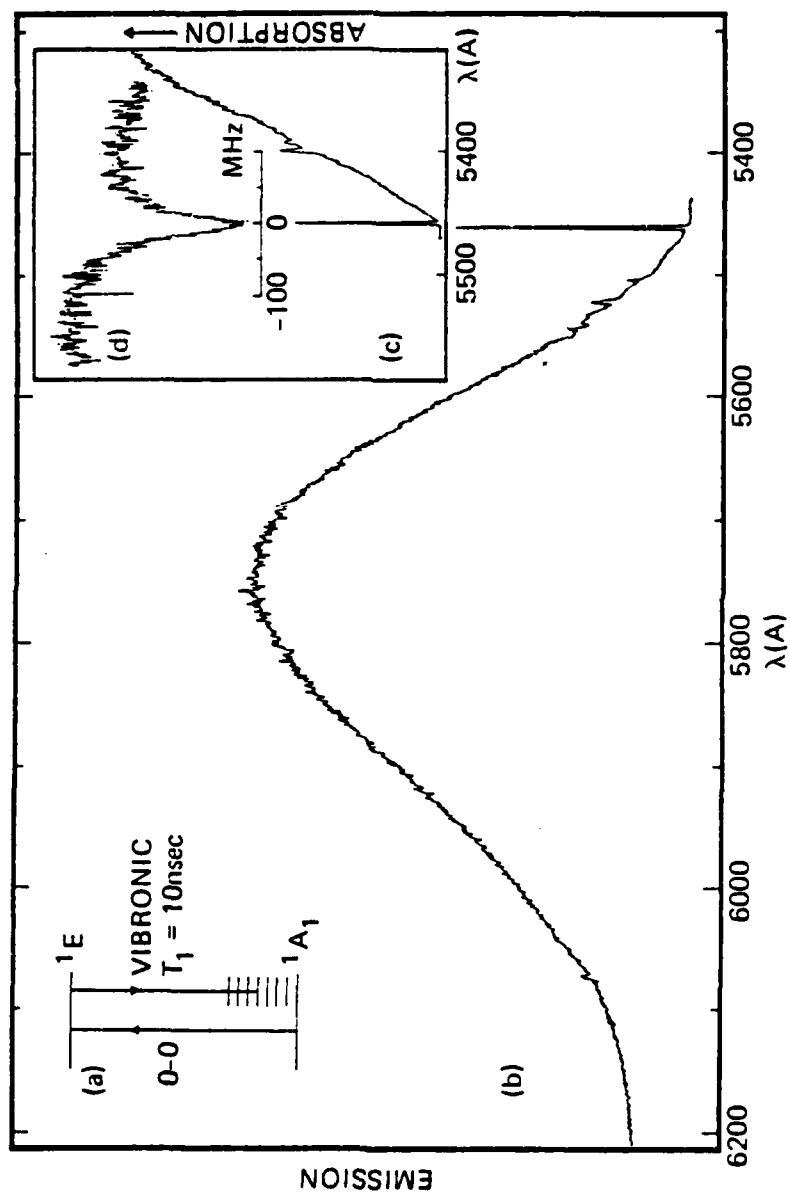


Figure 1

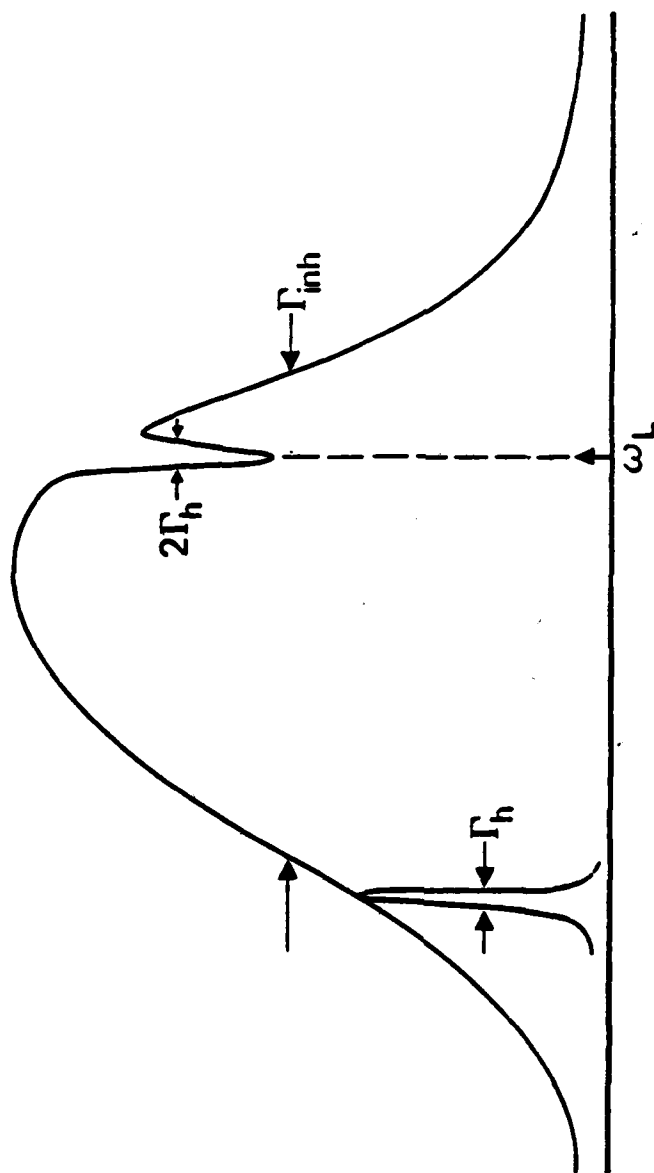


Figure 2

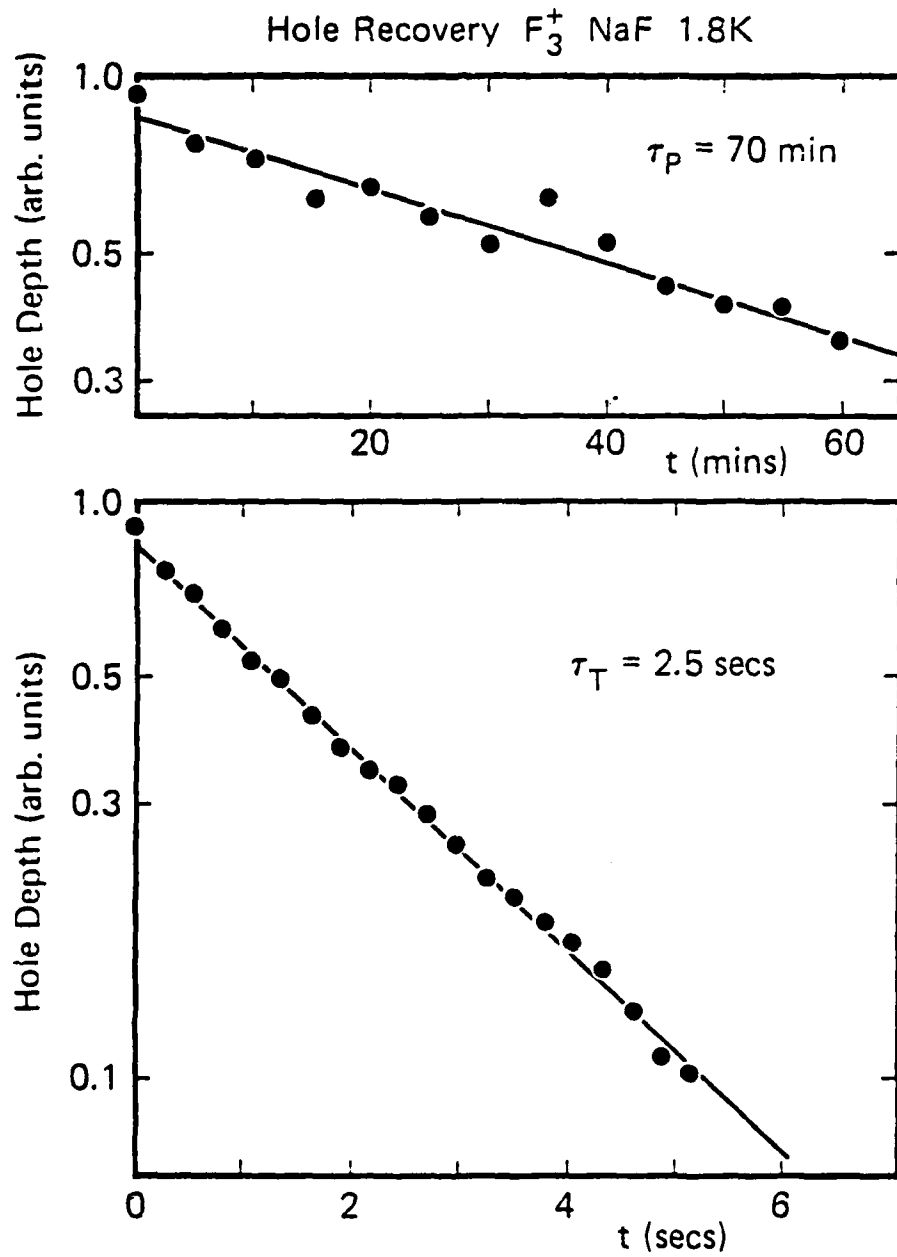


Figure 3

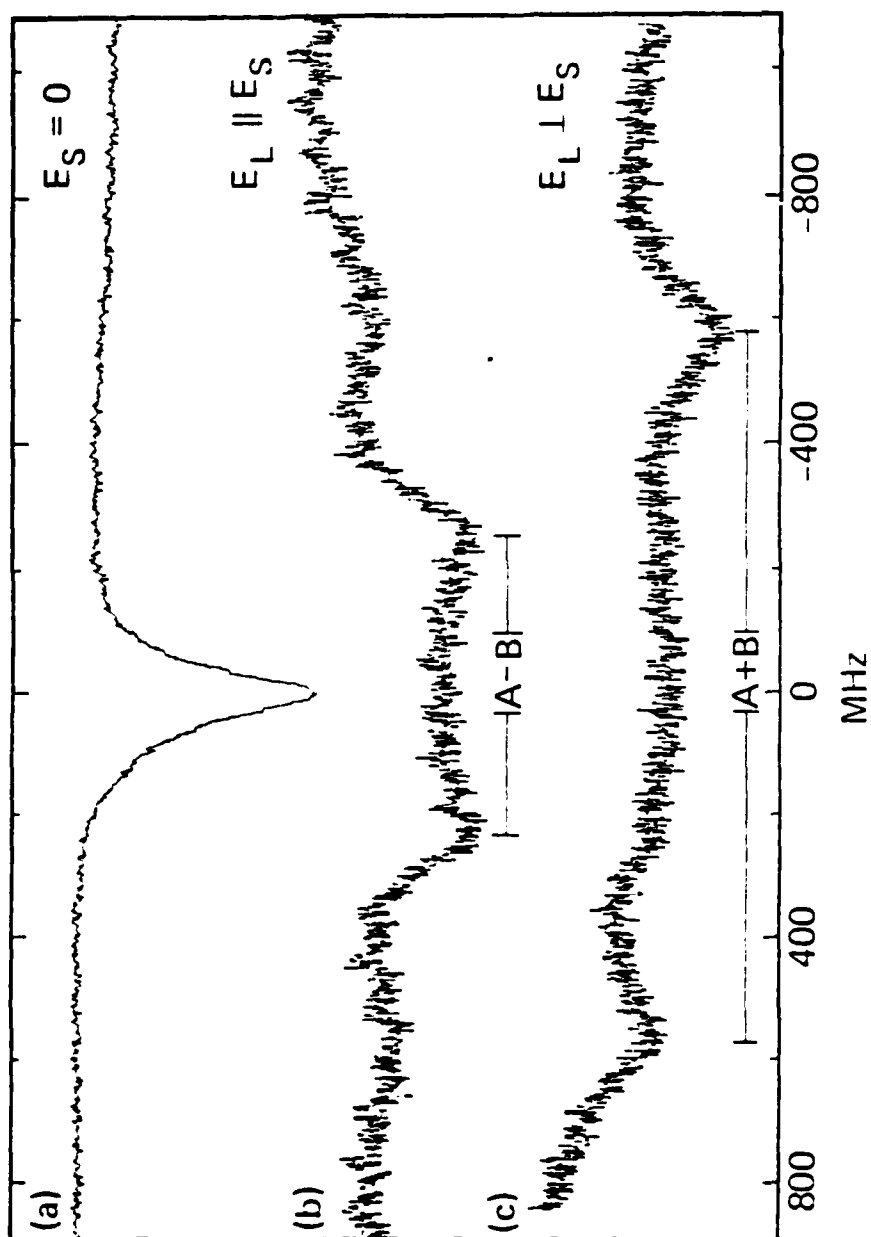


Figure 4

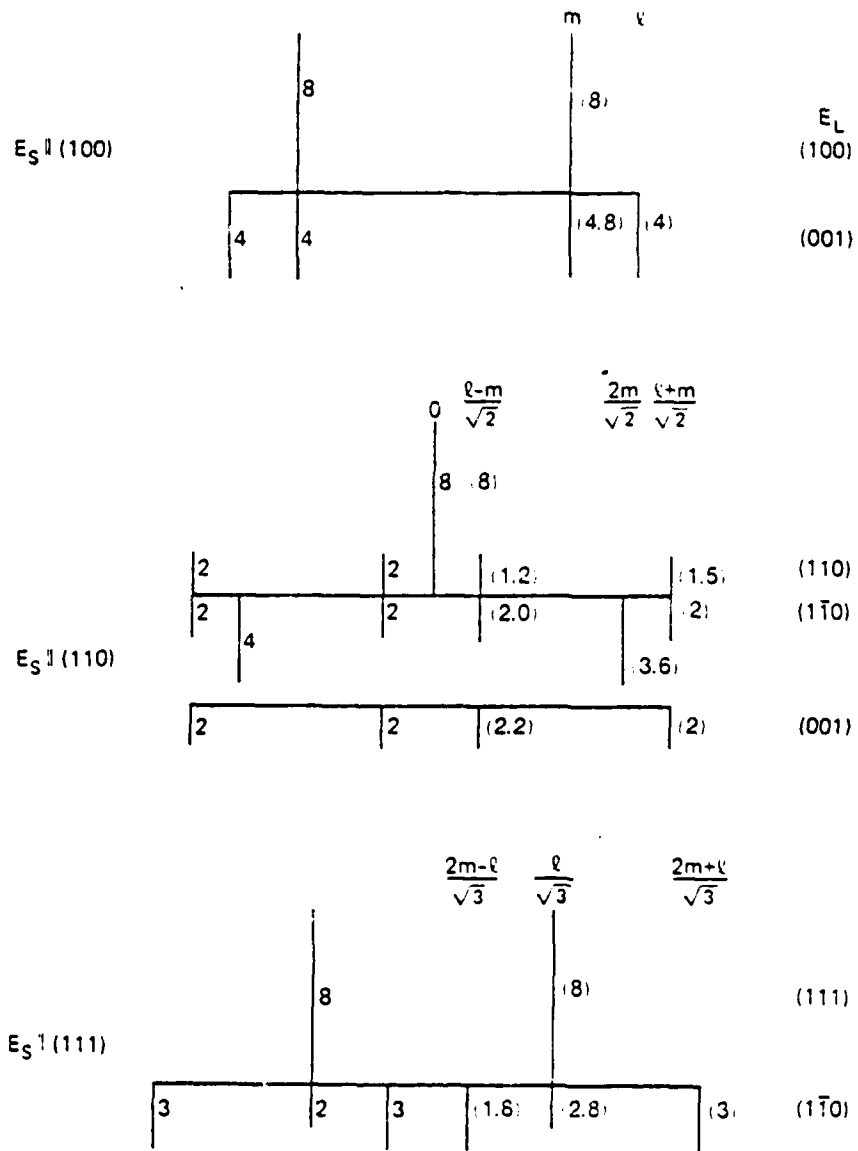


Figure 5

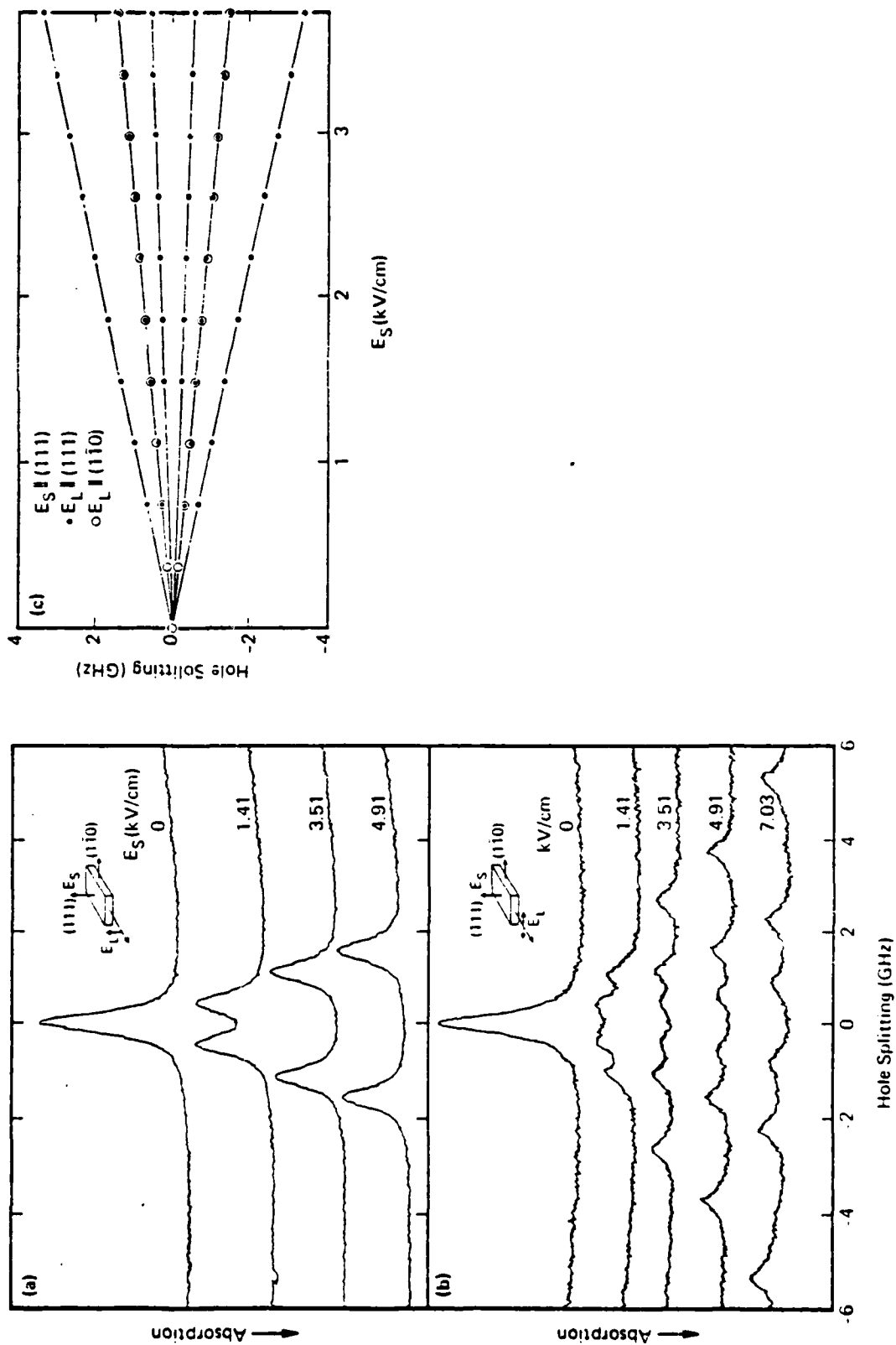


Figure 6

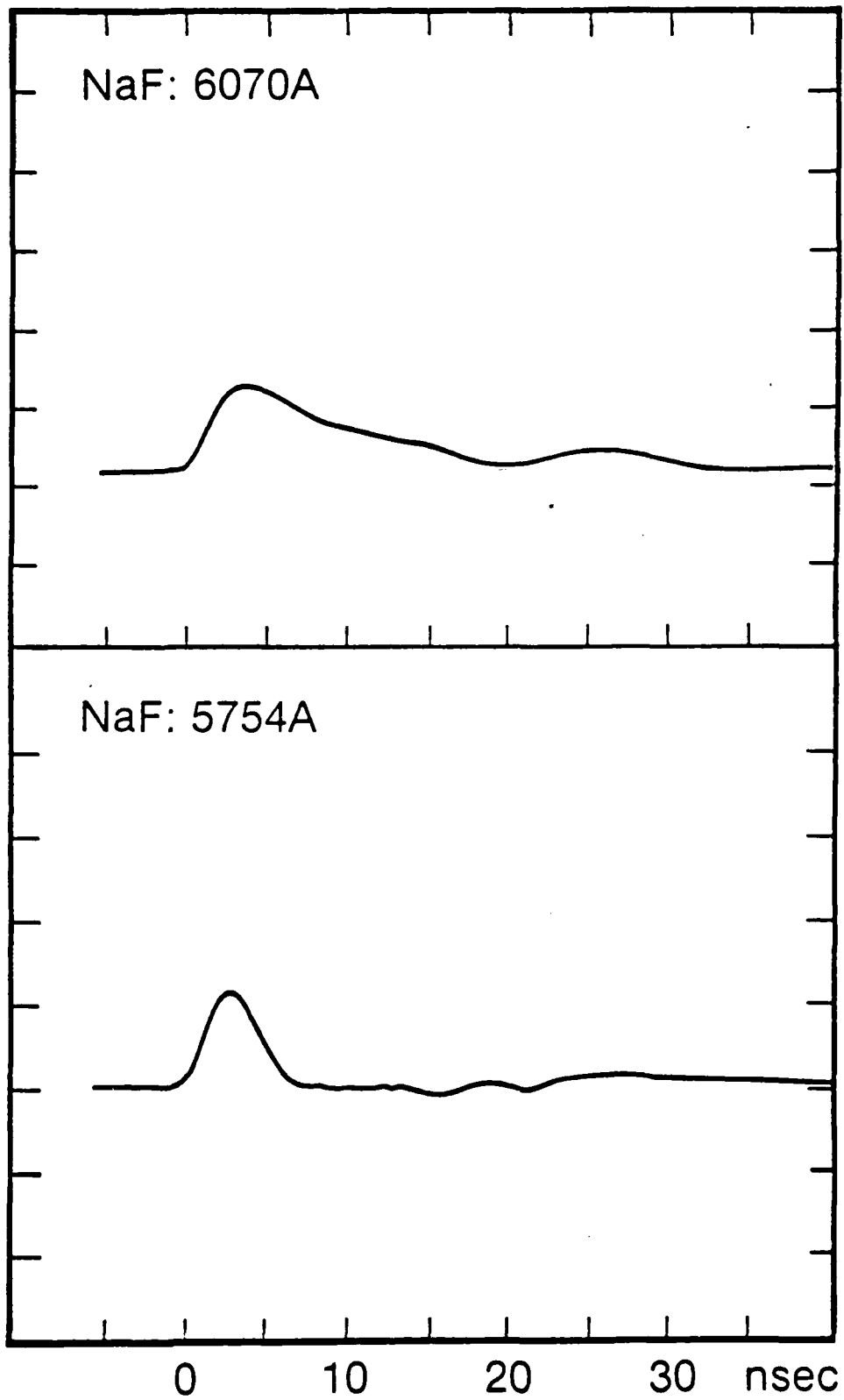


Figure 7

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